

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No. : 09/524,575 Confirmation No. 7265  
Applicant : Takuya HIRAMATSU et al.  
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DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Akira TAKAHASHI, declare and state:

(1) THAT I am a scientist who majored in adsorption and catalyst technology relating to exhaust gas purification and received a Ph.D. degree from the University of Michigan, Ann Arbor, Michigan, U.S.A.

(2) THAT I am one of the inventors of the subject matter claimed in U.S. Patent Nos. 5,118,644; 5,229,046; 5,178,809; 5,691,261; 5,804,521; 5,884,473; 6,029,441; 6,051,205; 6,139,808; 6,171,557; 6,207,604; 6,294,150; 6,350,416; 6,500,392 and 6,517,785.

(3) THAT I am one of the inventors of the method for exhaust gas purification described and claimed in U.S. Patent Application

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Serial No. 09/524,575 and am aware that the claims of the application have been rejected as being obvious over certain prior U.S. patents.

(4) THAT to demonstrate that the system for purification of exhaust gas from an internal combustion engine of the present invention in which an adsorbent zone and a catalyst zone are positioned in an in-line exhaust pipe of an internal combustion engine with the adsorbent zone being upstream of the catalyst zone with respect to flow of said exhaust gas, and in which the adsorbent contains an H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 100 or more and further contains at least one noble metal selected from Pt, Pd and Rh as a catalyst component, provides unexpected results as compared to the apparatus for treating engine exhaust gases of WO 94/11623 and, particularly, as compared to the apparatus for treating engine exhaust gas of Examples 2 and 3 of WO 94/11623, which includes first and second catalyst zones and an adsorbent zone between them, wherein the first and second catalyst zones are in heat transfer relation to one another via a crossflow monolith on which the catalysts are coated, the following tests (identified hereinafter as Additional Examples 1 and 2) were conducted under my direction and supervision.

(A) Preparation of Adsorbent-catalysts

Two Adsorbent-catalysts A1 and A2 for use in Additional Examples 1 and 2, respectively, were prepared in the same manner as that used

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to prepare Adsorbent-catalyst C as described in the specification of the present application except that  $\beta$ -zeolites having  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios of 110 and 210, respectively, were used to prepare the adsorbent. The  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of the  $\beta$ -zeolite used in Examples 2 and 3 of WO 94/11623 is 180.

The amount of the  $\beta$ -zeolite of Adsorbent-catalyst A1 and of Adsorbent-catalyst A2 carried on the honeycomb structure was 198 g. This amount is the amount of  $\beta$ -zeolite of the adsorbent of Examples 2 and 3 of WO 94/11623 and was calculated from the data of Examples 1-3 of WO 94/11623 as follows:

Ratio of  $\beta$ -zeolite actually loaded =

Weight of  $\beta$ -zeolite in zeolite sol ÷

(Weight of  $\beta$ -zeolite in sol + weight of solid silica in silica sol)

Total amount of  $\beta$ -zeolite actually loaded =

Amount of  $\beta$ -zeolite per unit x volume of honeycomb monolith  
x ratio of  $\beta$ -zeolite in the zeolite sol

The  $\beta$ -zeolite used in Example 1 of WO 94/11623 is 650 g, and the solid content of silica used in silica sol whose solid content is 22.8% is 315 g of the sol, and 635 g of deionized water

According to the description of Example 2, the honeycomb monoliths have been prepared in the same manner as those in Example 1.

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Thus, Examples 2 and 3 were calculated as shown below:

$$650 \text{ g} ) (650 + (315 \times 0.228)) = 0.9005$$

The amount of the  $\beta$ -zeolite loaded can be given as shown below:

$$2 \text{ g/in}^3 \text{ H } 110 \text{ in}^3 \times 0.9005 = 198 \text{ g}$$

#### (B) Purification System

The purification system used in Additional Examples 1 and 2 is the system used in Example 1 of the present application (refer to Table 2 on page 26) except that Adsorbent-catalyst A1 and Adsorbent-catalyst A2 were used in place of Adsorbent-catalyst C used in Example 1. The total amount of catalyst (Pt, Pd and Rh) of the purification system of Additional Examples 1 and 2, i.e., Catalyst A + Adsorbent-catalyst A1 or Adsorbent-catalyst A2 +, Catalyst B, was 5.05 g and 4.95 g, respectively. These amounts are comparable to an amount of catalyst of 4.99 g used in Example 2 of WO 94/11623 and a total amount of catalyst of 6.65 g used in Example 3 of WO 94/11623.

The amount of catalyst used in Example 2 of WO 94/11623 was calculated from the data of Example 2 as follows:

$$\begin{aligned} &\text{Volume of crossflow monolith (15.24 cm x 15.24 cm x 15.24 cm =} \\ &3.54 \text{ L) x precious metal content (1.41 g/L) = 4.99 g.} \end{aligned}$$

The amounts of catalyst used in Additional Examples 1 and 2 and

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Examples 2 and 3 of WO 94/11623 are identified in the following table:

	Total amount of catalyst used (g)	Catalyst carried on zeolite (g)	Catalyst carried on carrier other than zeolite (g)
Add. Exam. 1	5.05	1.66	1.13 + 2.26
Add. Exam. 2	4.95	1.56	1.13 + 2.26
Exam. 2 of WIPO	4.99	0	4.99
Exam. 3 of WIPO	6.65	0	4.99 + 1.66

Prior to use in the purification systems of Additional Examples 1 and 2, Adsorbent-catalysts A1 and A2 were subjected to the same thermal treatment as that of Adsorbent A as described in the specification of the present application on page 13, lines 4-11.

(C) Emission Test

Emission tests using the purification systems of Additional Examples 1 and 2 were conducted according to the procedure described in the specification of the present application on page 23, line 22, to page 24, line 20, using the system components described for Example 1, except that the Adsorbent-catalysts A1 and A2 were used in place of Adsorbent-catalyst C used in Example 1. The results of the emission tests are shown in the following Table together with the results of the

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emission test of the exhaust treatment systems of Examples 2 and 3 of WO 94/11623 as described in Table I and Table II.

	Zeolite (SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> )	Amount Used (g)	Type of System	Total amt. of Catalyst (g)	Emission in FTP Test (g/mile)	Purification Ratio (%)
Add. Ex. 1	β(110)	197	In-line	5.05	0.061	70
Add. Ex. 2	β(210)	195	In-line	4.95	0.052	77
Ex. 2 WO	β(180)	198	Cross	4.99	0.161	51
Ex. 3 WO	β(180)	198	Cross	4.99	0.065	62

In the above table, the purification ratio in Examples 2 and 3 of WO 94/11623 was calculated by using the data where no adsorbent was used and that wherein adsorbent was used. On the other hand, in the Additional examples, the purification ratio was calculated by using the data obtained in the case where no adsorbent/catalyst was used and that wherein the adsorbent/catalyst was used.

#### (D) Discussion

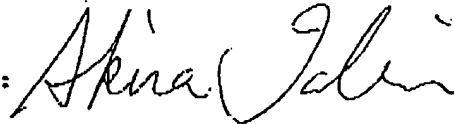
As can be seen, the system for purification of exhaust gas of the present invention provides unexpectedly superior results to the exhaust gas purification system of Examples 2 and 3 of WO 94/11623.

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(5) THAT all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and that further these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent resulting therefrom.

Signed this 18<sup>th</sup> day of February, 2005.

Signed:



Name: Akira TAKAHASHI